

ALUMINUM NITRIDE SINTERED BODY AND METHOD OF MANUFACTURING THE  
SAME

2. Claims

1. An aluminum nitride sintered body containing 0.001 to 0.2% by weight of carbon and having a thermal conductivity of 150 W/mk or more.

2. The aluminum nitride sintered body according to claim 1, containing any of the IIa group and IIIa group metals in an amount of 0.01 to 1.0% by weight in terms of oxide, containing 0.001 to 1.0% by weight of oxygen, and having a thermal conductivity of 180 W/mk or more.

3. A method of manufacturing an aluminum nitride sintered body, wherein the method involves mixing an aluminum nitride powder with any fatty acid salt of the IIa group and IIIa group metals in an amount of 0.01 to 1.0% by weight in terms of said metal oxide, forming the resulting blend, and subsequently sintering the compact in a non-oxidizing atmosphere at 1,500°C to 2,200°C.

3. Detailed Description of the Invention

[Field of the Invention]

The present invention relates to an aluminum nitride

sintered body and a method of manufacturing the same, and more specifically to a method of manufacturing a densified aluminum nitride sintered body excellent in practical properties such as thermal conductivity, insulating properties and dielectric constant.

[Prior Art]

Recent LSI shows a remarkable progress and has significant improvements in integration. To this contributes the improvement of IC chip sizes as well, and heat release per package is increasing as IC chip sizes are improved. Because of this, special emphasis has been placed on heat radiation properties of substrate materials. In addition, the thermal conductivity of an alumina sintered body that is conventionally used as the IC substrate is insufficient in heat radiation, and thus the body cannot almost deal with the increase in heat release of the IC chip. Hence, a beryllia substrate of high thermal conductivity has been studied as an alternative to the alumina substrate, however, beryllia is highly toxic and thus requires difficult handling.

Aluminum nitride (AlN) is essentially a material of high thermal conductivity and high insulating properties and also is nontoxic, and therefore is receiving attention as an insulating material or package material in the semiconductor industry.

[Problems to be Solved by the Invention]

As described above, aluminum nitride as a single crystal is theoretically a material of high thermal conductivity and high insulating properties. However, production of a sintered body from an aluminum nitride powder, after powder forming, leads to an aluminum nitride sintered body obtained by sintering which has a relative density (on the basis of the theoretical density of aluminum nitride of  $3.26 \text{ g/cm}^3$ ) of at most 70 to 80%, although depending on the sintering conditions, and which contains a large number of pores, because of the sintering ability of an aluminum nitride powder itself being not good.

Meanwhile, the heat conduction mechanism of an insulating ceramics like an aluminum nitride sintered body primarily consists of phonon conduction, and defect factors such as pores and impurities cause phonon scattering, thereby leading to a sintered body with a low level of thermal conductivity.

In order to obtain a densified aluminum nitride sintered body of good thermal conductivity, addition of a variety of sintering aids to an aluminum nitride powder followed by hot pressing or normal-pressure sintering has been tried. A fairly good sintered body was provided by, for example, a method that involves addition of calcium oxide (CaO), barium oxide (BaO), strontium oxide (SrO) and the like to an aluminum nitride powder, and then sintering (JP Kokoku Sho 58-49510). This method provides a sintered body having a thermal conductivity of 0.10 to 0.13 cal/cm·sec·deg (42 to 54 W/m·k) (room temperature) and

a relative density of 98% or more. However, this level of thermal conductivity does not suffice to deal with an increase in heat radiation caused by integration improvement of IC and LSI in the future.

On the other hand, for the purpose of obtaining a densified, high-strength aluminum nitride sintered body, an attempt to add  $Y_2O_3$ ,  $SiO_2$  and the like to an aluminum nitride powder has been made (JP Kokoku Sho 56-9475) to thereby obtain a sintered body with a relative density of 98% or more, but with a thermal conductivity of a low level, less than  $0.07 \text{ cal/cm} \cdot \text{sec} \cdot \text{deg}$  ( $29 \text{ W/m} \cdot \text{k}$ ).

The objects of the present invention are to provide a densified aluminum nitride sintered body excellent in practical properties such as thermal conductivity, insulating properties and dielectric constant so as to be suitably usable as a semiconductor insulating material or package material in the future, and a manufacturing method thereof.

[Means for solving the Problems]

The present invention for attaining the above-mentioned object comprises an aluminum nitride sintered body containing 0.001 to 0.2% by weight of carbon and having a thermal conductivity of  $150 \text{ W/mk}$  or more. In particular, a sintered body containing any of the IIa group and IIIa group metals in an amount of 0.01 to 1.0% by weight in terms of oxide and containing 0.001 to 1.0% by weight of oxygen has a thermal conductivity of  $180 \text{ W/mk}$  or

more.

A method of manufacturing such an aluminum nitride sintered body involves mixing an aluminum nitride powder with any fatty acid salt of the IIa group and IIIa group metals in an amount of 0.01 to 1.0% by weight in terms of said metal oxide, forming the resulting blend, and subsequently sintering the compact in a non-oxidizing atmosphere at 1,500°C to 2,200°C.

A sintered body of the present invention has a carbon content of 0.001 to 0.2% by weight. Rendering the carbon content to be less than 0.001% by weight drops the thermal conductivity, whereas when the content exceeds 0.2% by weight, the densification of the sintered body is insufficient. For making carbon remain, an aluminum nitride compact is required to contain a material to be a carbon source. Supply sources of carbon can be the forming aid, carbon to be added, carbon invaded from the atmosphere, and sintering aid. The inventors have found out that use of a fatty acid compound of the IIa and IIIa elements to be a sintering aid particularly leads to a preferable result.

While the action of carbon is not elucidated, it seems to be effective in reducing the amount of oxygen in AlN.

The amount of the IIa and IIIa elements is from 0.01 to 1.0% by weight in terms of oxide. Making the amount less than 0.01% by weight results in insufficient densification, whereas rendering the amount to exceed 1.0% by weight drops the thermal conductivity. The amount of oxygen in a sintered body is from

0.001 to 1.0% by weight. When the amount exceeds 1.0% by weight, the thermal conductivity is low, and controlling the amount to be less than 0.001% by weight is difficult.

Of the IIa or IIIa group elements as the sintering aid, hydrides, carbonates, etc. as well as oxides are usable, with fatty acid salts being particularly preferable. The inventors used various fatty acid salts of stearic acid, palmitic acid, etc. to obtain good results. The method of producing the sintered bodies can be in accordance with the usual method of manufacturing aluminum nitride.

#### [Examples]

The present invention will hereinafter be described in detail by means of examples.

#### Example 1

To a high-purity AlN powder (specific surface area  $2 \text{ m}^2/\text{g}$ ) having a mean particle diameter of  $1.0 \mu$  or less and having an oxygen content of 1.0% by weight were added a stearic acid compound of the IIa or IIIa group, the amount of addition being indicated in Table 1 in terms of oxide, and 5% by weight of PVB. The resulting material was blended in ethanol for 10 hours using a ball mill and molded and then sintered for two hours in a nitrogen flow at  $1,900^\circ\text{C}$ .

The relative density, thermal conductivity, degree of weight increase by PCT ( $120^\circ\text{C}$ , 100% RH, 100 H) of the sintered body thus obtained are given in Table 1 below.

The results indicated in this Table show that the properties of aluminum nitride sintered bodies of the present invention are excellent.

#### Example 2

No. 3 composition of Example 1 in Table 1 was employed except using palmitic acid instead of stearic acid. Similar results were obtained.

Table 1

No.	IIa, IIIa group element	In terms of oxide (wt%)	Relative density	Amount of carbon remaining (wt%)	Thermal conductivity (W/mk)	Weight increase (mg/cm <sup>2</sup> )	Amount of oxygen (wt%)
Example	1	Y	0.2	100	0.02	0.02	0.2
	2	Y	0.4	100	0.04	0.02	0.2
	3	Y	0.8	100	0.10	0.02	0.1
	4	Ga	0.6	100	0.04	0.03	0.1
	5	Sc	0.6	100	0.05	0.04	0.1
	6	Ca	0.6	100	0.05	0.03	0.1
	7	Mg	0.8	100	0.05	0.03	0.1
Comparative Example	8	Y <sub>2</sub> O <sub>3</sub>	0.6	99	0.001	0.03	0.3
	9	Y	1.2	100	0.05	0.10	0.1
	10	-	-	94	< 0.001	0.03	1.2



[Effect of the Invention]

As discussed thus far, the present invention is a highly reliable aluminum nitride sintered body of high thermal conductivity, which can be relatively readily produced. Also, an aluminum nitride sintered body of the present invention has properties suitable for IC substrates, heat radiating plates, structural materials, or the like, and is highly practical.

(Attached Sheet)

(1) Claims are amended in the following.

"2. Claims

1. An aluminum nitride sintered body containing 0.001 to 0.2% by weight of carbon, containing any of the IIa group and IIIa group metals in an amount of 0.05 to 0.8% by weight in terms of oxide, containing 0.001 to 1.0% by weight of oxygen, and having a thermal conductivity of 180 W/mk or more.

2. A method of manufacturing an aluminum nitride sintered body, wherein the method involves mixing an aluminum nitride powder with any fatty acid salt of the IIa group and IIIa group metals in an amount of 0.05 to 0.8% by weight in terms of said metal oxide, forming the resulting blend, and subsequently sintering the compact in a non-oxidizing atmosphere at 1,500°C to 2,200°C."

(2) The expression "having a thermal conductivity .... In particular, a sintered body" in the specification, page 4, lines 21 to 22 is deleted.

(3) The expression "0.01 to 1.0" in the specification, page 4, lines 23 to 24 and page 5, line 5 is amended to "0.05 to 0.8".

(4) The expression "0.01 to 1.0" in the specification, page 6,

line 1 to 2 is amended to "0.05 to 0.8".

(5) The expression "0.01" in the specification, page 6, line 1 is amended to "0.05".

(6) The expression "1.0" in the specification, page 6, line 3 is amended to "0.8".

(7) Table 1 in the specification, page 8 is amended as below.

[Table 1]

No.	IIa, IIIa group element	In terms of oxide (wt%)	Relative density	Amount of carbon remaining (wt%)	Thermal conductivity (W/mk)	Weight increase (mg/cm <sup>2</sup> )	Amount of oxygen (wt%)
Example	1	Y	0.4	100	0.04	0.02	0.2
	2	Y	0.8	100	0.10	0.02	0.1
	3	Gd	0.6	100	0.04	0.03	0.1
	4	Sc	0.6	100	0.05	0.04	0.1
	5	Ca	0.6	100	0.05	0.03	0.1
	6	Mg	0.6	100	0.05	0.03	0.1
Comparative Example	7	Y <sub>2</sub> O <sub>3</sub>	0.6	99	< 0.001	0.03	0.3
	8	Y	1.2	100	0.05	0.10	0.1
	9	-	-	94	< 0.001	0.03	1.2